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AUGUST 1961

# FREE RADICALS
# BIBLIOGRAPHY AND SURVEY OF PUBLICATIONS
# (up to 1959)

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Ordnance Management Structure Code No. 5610.11.703
BALLISTIC RESEARCH LABORATORIES

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Terminal Ballistics Laboratory



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A B E R D E E N   P R O V I N G   G R O U N D,   M A R Y L A N D

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GLavin/ACoates/JRakaczky/bjk
Aberdeen Proving Ground, Md.
August 1961

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ABSTRACT

A survey has been made of the free radical literature published up to and including the year 1959. Approximately 2200 references are presented in the form of the title of the paper, author, and a brief description of the work where the title is not self-explanatory.

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## INTRODUCTION

In the course of the evolution of chemical philosophy the term radical has been used in varying senses, and survivals of earlier significance persist in present day usage. The original concept long antedated clear-cut ideas on molecular structure and the nature of valence forces. It is, perhaps, a fair approximation to state that the word radical originally signified an atomic aggregate or molecular fragment capable of surviving as an entity throughout a series of chemical transformations.

Free radicals, as usually defined in terms of modern valency concepts, are individual atoms of groups of atoms of electronic configuration such that it includes at least one unpaired electron. Such a definition is broad enough to include atoms (other than those of the noble gases), polyatomic molecular fragments such as the long-lived triaryl methyl radicals described by Gomberg, as well as the short-lived alkyl radicals of the Paneth type.

Free radicals thus are fragments of matter which for the most part have only a fleeting existence. They are present in an ordinary flame, in an electric arc, in the atmosphere in the stars and even in the cold interstellar dust. These highly reactive fragments can initiate and sustain a chain reaction among comparatively inactive substances. A relatively small number of free radicals (as few as one per thousand) can maintain a chain reaction. If we understood their behavior fully we would have a master key to the chemistry of the universe.

This survey covers the period up to and including 1959. The bibliography contains approximately 2200 items each of which consists of the author, subject, journal reference and a short abstract if the title of the paper is not self-explanatory. The references are listed alphabetically by the last name of the senior author. An author index also is included. This bibliography was compiled during the course of free radical investigations being conducted at these Laboratories. It is our wish that it will be as useful to others as it has been to us. It is also hoped that it will act as a catalyst in furthering future free radical research.

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$\text{Me} + \text{AcMe} = CH_4 + CH_2\text{COMe}$
$\text{Me} + CH_2\text{COMe} = \text{EtCOMe}$
Accounted for approx. 95% of the methyl radicals disappearing during the photolysis. No evidence for the reaction
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$OH + OH + M = H_2O_2 + M$
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Discharge of a condenser between W-electrodes in a quartz tube yields flashes of 1000 Joules at a duration of $10^{-4}$ sec. The method is also suitable to study explosions because a homogeneous ignition takes place over the entire length of the tube. The reactions and lifetimes of atoms and radicals Cl, Br, I, ClO, HS, CS, $CH_2$, $CH_3$, $CH_3CO$, NH, OH, $C_2$ can be followed by photographing their absorption spectra by means of timed secondary flashes.

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C. A., 28, 2672 (1934).
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C. A., 27, 5714 (1933).
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VIII. Comparison of the Thermal and Electrical Decomposition of Organic Compounds into Free Radicals.
J. Am. Chem. Soc., 56, 1311-13 (1934).
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IX. The Combination of Methyl Groups with Metallic Mercury.
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C. A., 28, 6697 (1934).
Thermal decomposition of propane or butane.

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The Thermal Decomposition of Organic Compounds from the Standpoint of Free Radicals.
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The $CH_2$ radical reacts with the metals: Te, Se, As, and Sb, but not at all with Zn, Cd, Hg, Tl, Pb, or Bi. The methyl group, $CH_3$, reacts with all the metals listed, thus affording a conclusive test as to whether $CH_2$ or $CH_3$ have been formed.

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C. A., 29, 723 (1935).
No HCH Te is formed, thus proving the absence of $CH_2$ radicals in the gases leaving the furnace. No hydrides of Te were detected in the liquid air trap indicating the absence of atomic hydrogen among the fragments leaving the furnace. The experiments indicate that $CH_4$ undergoes a primary dissociation into $CH_3$ and H; the H disappears before reaching the mirror, either on the wall or by reaction with the $CH_4$ to form $CH_3$ and $H_2$.

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Thermal Decomposition of Organic Compounds from the Standpoint of Free Radicals.
XIII. The Decomposition of Ethyl Nitrite.
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C. A., 29, 723 (1935).
When pure $EtNO_2$ vapor at low pressure is decomposed in a flowing system, cold metallic mirrors are not affected by the gases leaving the furnace. On the other hand, if the $EtNO_2$ is diluted with an inert gas, mirrors are readily removed, even when the furnace is at temperatures as low as 425°C. This behavior may be explained by assuming that the interaction of a free radical with $EtNO_2$ results in the production of molecules only. The activation energy of the primary dissociation of $EtNO_2$ is $34.3 \pm 3$ cal.

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The Role of Free Radicals in Elementary Organic Reactions.
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The relationship between molecular structure and reactivity.

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The mechanism of the $C_2H_6$ decomposition is discussed. A modification of the original scheme by Kuchler and Thiele is examined in detail.

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The free radical theory is discussed in relation to the thermal decomposition of pure organic compounds.

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Me azide was passed through a gravity tube held at 900° at a pressure of 0.1 mm. The products were frozen out. A mechanism for the reaction is proposed.

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Relative Activation Energies of Removal of Primary, Secondary and Tertiary Hydrogen Atoms by Methyl Radicals.
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Reactions of Oxygen and Hydrogen at Low Pressures.
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